

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of

Atty Dkt. 608-312

ELLIS et al

C# M#

Serial No. 09/964,849

TC/A.U.

1625

Filed: September 28, 2005

Examiner: Oh, T.V.

Date: November 1, 2005

Title: OXIDATION PROCESS FOR THE PRODUCTION OF ALKENES AND CARBOXYLIC ACIDS

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

RESPONSE/AMENDMENT/LETTER

This is a response/amendment/letter in the above-identified application and includes an attachment which is hereby incorporated by reference and the signature below serves as the signature to the attachment in the absence of any other signature thereon.

☐ **Correspondence Address Indication Form Attached.**

Fees are attached as calculated below:

Total effective claims after amendment	58	minus highest number			
previously paid for	58	(at least 20) =	0	x \$50.00	\$0.00 (1202)/\$0.00 (2202) \$ 0.00

Independent claims after amendment	3	minus highest number			
previously paid for	3	(at least 3) =	0	x \$200.00	\$0.00 (1201)/\$0.00 (2201) \$ 0.00

If proper multiple dependent claims now added for first time, (ignore improper); add					
					\$360.00 (1051)/\$180.00 (2051) \$ 0.00

Petition is hereby made to extend the current due date so as to cover the filing date of this					
paper and attachment(s)					
					One Month Extension \$120.00 (1251)/\$60.00 (2251)
					Two Month Extensions \$450.00 (1252)/\$225.00 (2252)
					Three Month Extensions \$1020.00 (1253)/\$510.00 (2253)
					Four Month Extensions \$1590.00 (1254)/\$795.00 (2254) \$ 0.00

Terminal disclaimer enclosed, add					\$130.00 (1814)/\$65.00 (2814) \$ 0.00
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☐ Applicant claims "small entity" status. ☐ Statement filed herewith

Rule 56 Information Disclosure Statement Filing Fee					\$180.00 (1806) \$ 0.00
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Assignment Recording Fee					\$40.00 (8021) \$ 0.00
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Other: Appeal Brief					\$ \$500.00
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TOTAL FEE ENCLOSED \$ 500.00

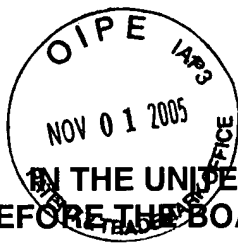
The Commissioner is hereby authorized to charge any deficiency, or credit any overpayment, in the fee(s) filed, or asserted to be filed, or which should have been filed herewith (or with any paper hereafter filed in this application by this firm) to our Account No. 14-1140. A duplicate copy of this sheet is attached.

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NIXON & VANDERHYE P.C.

By Atty: Leonard C. Mitchard, Reg. No. 29,009

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

In re Patent Application of

ELLIS et al

Atty. Ref.: 608-312

Serial No. 09/964,849

TC/A.U.: 1625

Filed: September 28, 2001

Examiner: Oh, T.V.

For: OXIDATION PROCESS FOR THE PRODUCTION OF ALKENES AND
CARBOXYLIC ACIDS

November 1, 2005

Mail Stop Appeal Brief - Patents
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

APPEAL BRIEF

Sir:

Applicant hereby appeals to the Board of Patent Appeals and Interferences from
the last decision of the Examiner.

11/02/2005 JADD01 00000011 09964849

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(I) REAL PARTY IN INTEREST

The real party in interest is BP Chemical Limited, a corporation of the country of the United Kingdom.

(II) **RELATED APPEALS AND INTERFERENCES**

There are no related appeals or interferences.

(III) STATUS OF CLAIMS

Claims 1-26 are pending and are rejected. Claims 1-26 are appealed. No claims have been substantively allowed.

(IV) STATUS OF AMENDMENTS

An amendment has been submitted subsequent to final rejection. The amendment, submitted on June 23, 2005, clarified the dependency of claim 5. In the Advisory Action mailed August 17, 2005, the Examiner has entered the amendment and has agreed that the amendment obviates the outstanding formal rejection.

(V) SUMMARY OF CLAIMED SUBJECT MATTER

In the following summary of the claimed subject matter, references in parenthesis are to support in the specification, the pages and lines referred to being of the specification. Claims 1-3 are the independent claims under appeal. Claims 4-26 are dependent, either directly or indirectly, on claim 1, 2 or 3.

As claimed in claim 1, the present invention is directed to a process for the oxidation of a C₂ to C₄ alkane to produce the corresponding alkene and carboxylic acid (page 3, beginning at line 3). The process comprises contacting in an oxidation reaction zone, the alkane, molecular oxygen-containing gas and the corresponding alkene in the presence of at least one catalyst active for the oxidation of the alkane to the corresponding alkene and carboxylic acid, to produce a product stream comprising alkene, carboxylic acid and water, and adjusting or maintaining the molar ratio of alkene produced in the oxidation reaction zone to carboxylic acid produced in the oxidation reaction zone by controlling the concentration of the alkene introduced in the oxidation reaction zone (page 3, lines 5-13).

Claim 2 is directed to an integrated process for the production of an alkyl carboxylate (page 4, beginning at line 20). The process comprises the steps of (a) contacting in an oxidation reaction zone a C₂ to C₄ alkane, a molecular oxygen-containing gas and the corresponding alkene in the presence of at least one catalyst active for the oxidation of the alkane to the corresponding alkene and carboxylic acid, to produce a product stream comprising alkene and carboxylic acid and water; adjusting or maintaining the molar ratio of alkene produced in the oxidation reaction zone to carboxylic acid produced in the oxidation reaction zone by controlling the concentration of alkene introduced in the oxidation reaction zone; and (b) contacting in a second

reaction zone at least a portion of each of the alkene and carboxylic acid produced in the first reaction zone, in the presence of at least one catalyst active for the production of alkyl carboxylate to produce the alkyl carboxylate (page 4, lines 22-29).

Claim 3 is directed to an integrated process for the production of an alkenyl carboxylate (page 5, beginning at line 2). The process comprises the steps of (a) contacting in an oxidation reaction zone a C₂ to C₄ alkane, a molecular oxygen-containing gas, and the corresponding alkene and optionally, water in the presence of at least one catalyst active for the oxidation of the alkane to the corresponding alkene and carboxylic acid, to produce a product stream comprising alkene and carboxylic acid and water; adjusting or maintaining the molar ratio of alkene produced in the reaction zone to carboxylic acid produced in the oxidation reaction zone by controlling the concentration of alkene introduced in the oxidation reaction zone; and (b) contacting in a second reaction zone at least a portion of each of the alkene and carboxylic acid produced in the first reaction zone and a molecular oxygen-containing gas, in the presence of at least one catalyst active for the production of alkenyl carboxylate to produce the alkenyl carboxylate (page 5, lines 4-17).

(VI) GROUND OF REJECTION TO BE REVIEWED ON APPEAL

The grounds of rejection to be reviewed on appeal are as follows. Claims 1, 4, 7, and 11-26 stand rejected under 35 U.S.C. §103 as allegedly unpatentable over U.S. Patent 5,162,578 to McCain in view of U.S. Patent 4,899,003 to Manyik et al. Claims 2, 3, 5, 6 and 8-10 stand rejected under 35 U.S.C. §103(a) as allegedly unpatentable over U.S. 3,458,406 to Fisher et al in view of Manyik et al.

(VII) ARGUMENT

The presently claimed invention requires that **both** an alkane, such as ethane, **and** an alkene, such as ethylene, are reacted with oxygen in the presence of a catalyst to produce **both** a carboxylic acid such as acetic acid **and** an alkene such as ethylene. The inventive concept underlying the present invention is the discovery that the molar ratio of carboxylic acid produced to alkene produced can be tailored by controlling the concentration of alkene reactant introduced in the oxidation reaction zone.

The cited art does not disclose or suggest the concept of adjusting or maintaining the molar ratio of alkene **produced** in the oxidation reaction zone to carboxylic acid **produced** in the oxidation reaction zone by controlling the concentration of alkene **introduced** into the oxidation reaction zone. McCain aims to provide a mixed catalyst composition which produces predominantly acetic acid from ethane and/or ethylene in a process which is **selective to acetic acid** (column 3, lines 7-8 and lines 50-54). McCain is **not** interested in ethylene as a product. McCain discloses that the solution for achieving selectivity to acetic acid is to use a mixture of two catalyst species (column 3, lines 58-61). At column 1, lines 45-48, McCain states that the prior art reference, US 4,524,236, does not suggest or disclose mixed catalyst compositions for selective acid production.

McCain does not specify adjustment of the molar ratio of alkene **produced** to carboxylic acid **produced** by controlling the concentration of alkene **introduced** into the reaction zone. This deficiency is not cured by Manyik. Manyik relates to a process for the conversion of ethane to ethylene (column 1, lines 6-7). Ethylene is not a feed component as required by the presently claimed invention. The process is multistage such that ethane is reacted with oxygen in the presence of a catalyst to produce at each

stage an output stream comprising ethylene, acetic acid, water and unreacted ethane and oxygen. The composition of the output stream is altered by adjusting the amount of **acetic acid and/or water in the stream** (col. 1, lines 32-48; claim 1). Manyik does not disclose or suggest adjusting the composition of the output product stream by adjusting the concentration of alkene introduced into the reaction zone. In fact, as noted earlier, in Manyik, ethylene is not a feed component, as required by the appealed claims. At col. 6, lines 6 1-63, Manyik states that the preferred catalysts for use in the invention are those described in US 4,524,236 (cf. column 1, lines 45-48 of McCain which state that US 4,524,236 does not suggest or disclose mixed catalyst compositions for selective acid production).

Referring again to Manyik, the Examiner draws attention at page 6 of the Action to the passage at column 3, lines 10-18 of Manyik, which reads as follows:

The mole ratio of ethylene to acetic acid defines the relative yield of these products. It will be described herein, process steps for changing this ratio for use in a commercial viable process according to the invention. Thus, a commercial process can be carried out to favor the production of ethylene at the expense of acetic acid or to favor the production of acetic acid at the expense of ethylene.

This passage in Manyik does not suggest the inventive concept of the present invention that the molar ratio of alkene in the product and acid in the product is controlled by adjusting the concentration of the alkene introduced in the oxidation reaction zone. Rather, Manyik discloses the removal of water as a way to adjust the "distribution of the production of products in favor of ethylene at the expense of acetic acid." (column 6 lines 20-26).

There would have been no motivation for one of ordinary skill to combine McCain and Manyik. The aims and processes of McCain and Manyik are different. McCain is

specifically directed to the production of only acetic acid (column 3, lines 7-8 and lines 50-54), i.e., McCain leads specifically **away** from producing both acetic acid and ethylene, which is the subject-matter of Manyik. Thus, there would have been no motivation for the skilled person to combine a disclosure of a process aimed at producing solely acetic acid with a process which is totally contrary to this aim, i.e., one that aims to reduce the production of acid by producing ethylene as well. This is corroborated by:

(a) the nature of the catalysts used in McCain and Manyik. Manyik states at column 6, lines 56-63 that the preferred catalysts for use in the process of Manyik are those of US 4,524,236 which relates to a process for the conversion of ethane to ethylene. These catalysts are stated by McCain as not being able to selectively produce acetic acid and therefore cannot achieve the invention of McCain (column 1, lines 45-48) (Manyik on the other hand states that the preferred catalysts for use in the invention are those described in US 4,524,236);

(b) any ethylene by-product produced in McCain is recycled to make more of the desired acetic acid (column 4, lines 30-34).

The Examiner states at page 7 of the Action that Manyik indicates that the addition of water results in high acetic acid selectivity whilst reducing the ethylene selectivity. From this, the Examiner concludes that the skilled person would be motivated to incorporate Manyik's ethane and water into the process of McCain to increase the selectivity to acid. Applicant's disagree.

Example 1 of McCain employs water (column 11, line 13) and two runs of the experiment are carried out. In Run A, the catalyst system of McCain's invention is employed whereas, in Run B, a comparative experiment is conducted using the

preferred catalyst system of Manyik (i.e. MoVNbSbCa — see column 7, line 20 of Manyik). In Run A, 63 mol% acid is obtained; in Run B only 32 mol% acid is obtained. Example 2 of McCain describes Run A which uses the catalyst invention of McCain and Run B which is a comparative experiment using the preferred catalyst of Manyik. Ethylene is reacted with oxygen to produce acetic acid and carbon oxides. In Run A, selectivity to acetic acid was 74 mol%, in Run B it dropped to the low value of 41 mol%. Thus, much poorer results are obtained by using the system of Manyik. It is also noted that ethylene is not a required feed component in McCain (as it is in the present invention). McCain, at column 1, lines 7-9, states that acetic acid may be produced by the catalytic oxidation of ethane or ethylene, or mixtures of ethane and ethylene. Thus, ethane may be used as the sole hydrocarbon feed component. The Examiner's attention is directed to Example 7 of McCain in which the oxidation of ethane alone was carried out.

McCain clearly leads one of ordinary skill **away** from employing the inferior process and teaching of Manyik. Based on this, there would have been no incentive for the skilled person to incorporate any teaching from Manyik to McCain to increase selectivity to acid because McCain is already an improved process for acid production over Manyik. Even if the skilled person did choose to add additional water, greater selectivity to acid may or may not be obtained in McCain because the nature of the catalyst in McCain is different to that of Manyik.

Furthermore, even if the addition of water did achieve further selectivity to acetic acid, this is irrelevant. An essential requirement of present claim 1 is that the ethylene concentration introduced is controlled so as to adjust the molar ratio of ethylene to acetic acid produced. McCain does not disclose or suggest this concept. Hence, a

combination of McCain and Manyik would not lead the skilled person to the present invention.

For the above reasons and contrary to the Examiner's assertions, the skilled person would have had no incentive to combine the teachings of McCain and Manyik and, even if that person did attempt such a combination, neither McCain nor Manyik discloses that by varying the ethylene concentration introduced, the product molar ratio may be adjusted/maintained at a predetermined value. Reversal of the obviousness rejection is accordingly respectfully requested.

Referring to the obviousness rejection over Fisher in view of Manyik, claims 2 and 3 are independent claims directed to integrated processes having, as a first step, the oxidation process of claim 1 to produce acetic acid and ethylene and reacting these products in a second step to produce either an alkyl carboxylate, such as ethyl acetate (claim 2) or an alkenyl carboxylate, such as vinyl acetate (claim 3). Fisher relates to the purification of vinyl acetate by removal of methyl acetate and ethyl acetate impurities. The vinyl acetate is produced by reacting ethylene, acetic acid and oxygen in the presence of a catalyst.

As noted above, Manyik is completely silent on any suggestion of controlling ethylene reactant concentration to tailor acid and ethylene product ratio. The Examiner states on page 7 of the Action that Fisher has focused on the production of the final products, alkyl carboxylate and alkenyl carboxylate by using ethylene and acid. This is incorrect. Fisher is directed to the purification of vinyl acetate to remove methyl and ethyl acetate impurities (Col. 1 lines 45-52 lines 65-67; Col 2 lines 20-23; Col. 4 line 49). Thus, the only final product in Fisher is vinyl acetate.

Fisher relates to vinyl acetate. Manyik relates to ethylene and acetic acid. The two references therefore relate to completely different technical fields. In addition, Fisher is only concerned with vinyl acetate that has already been produced, and is not concerned at all with processes on how to make vinyl acetate. Thus, contrary to the Examiner's contentions, the skilled person would have had no motivation to combine the disclosures of Fisher and Manyik. Furthermore, as neither Fisher nor Manyik provides any disclosure or suggestion of how to tailor the acetic acid to ethylene product ratio via controlling ethylene reactant concentration, and as neither reference relates to the production of an alkyl carboxylate, the skilled person could not have arrived at the subject-matter of claims 2, 3 or their dependent claims by a combination of Manyik and Fisher.

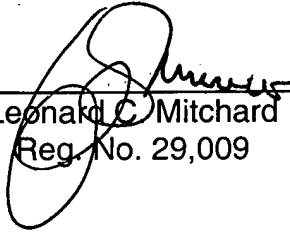
In light of the above, it is clear that one of ordinary skill would not have been motivated to arrive at the invention as claimed in the present application based on the combined disclosures of McCain and Manyik or Fisher and Manyik. Absent any such motivation, a *prima facie* case of obviousness has not been generated in this case.

Reversal of the obviousness rejections is respectfully requested.

Respectfully submitted,

NIXON & VANDERHYE P.C.

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(VIII) CLAIMS APPENDIX

1. A process for the oxidation of a C₂ to C₄ alkane to produce the corresponding alkene and carboxylic acid which process comprises:

contacting in an oxidation reaction zone, said alkane, molecular oxygen-containing gas and the corresponding alkene in the presence of at least one catalyst active for the oxidation of the alkane to the corresponding alkene and carboxylic acid, to produce a product stream comprising alkene, carboxylic acid and water; and

adjusting or maintaining the molar ratio of alkene produced in said oxidation reaction zone to carboxylic acid produced in said oxidation reaction zone by controlling the concentration of the alkene introduced in said oxidation reaction zone.

2. An integrated process for the production of an alkyl carboxylate which process comprises the steps:

(a) contacting in an oxidation reaction zone a C₂ to C₄ alkane, a molecular oxygen-containing gas and the corresponding alkene in the presence of at least one catalyst active for the oxidation of the alkane to the corresponding alkene and carboxylic acid, to produce a product stream comprising alkene and carboxylic acid and water; adjusting or maintaining the molar ratio of alkene produced in said oxidation reaction zone to carboxylic acid produced in said oxidation reaction zone by controlling the concentration of alkene introduced in said oxidation reaction zone; and

(b) contacting in a second reaction zone at least a portion of each of said alkene and carboxylic acid produced in the first reaction zone, in the presence of at least one catalyst active for the production of alkyl carboxylate to produce said alkyl carboxylate.

3. An integrated process for the production of an alkenyl carboxylate which process comprises the steps:

(a) contacting in an oxidation reaction zone a C_2 to C_4 alkane, a molecular oxygen-containing gas, and the corresponding alkene and optionally, water in the presence of at least one catalyst active for the oxidation of the alkane to the corresponding alkene and carboxylic acid, to produce a product stream comprising alkene and carboxylic acid and water; adjusting or maintaining the molar ratio of alkene produced in said reaction zone to carboxylic acid produced in said oxidation reaction zone by controlling the concentration of alkene introduced in said oxidation reaction zone; and

(b) contacting in a second reaction zone at least a portion of each of said alkene and carboxylic acid produced in the first reaction zone and a molecular oxygen-containing gas, in the presence of at least one catalyst active for the production of alkenyl carboxylate to produce said alkenyl carboxylate.

4. A process as claimed in claim 1, 2 or 3 in which the molar ratio of alkene to carboxylic acid produced in the oxidation reaction zone is in the range 10:1 to 1:10.

5. A process as claimed in claim 1, 2 or 3 in which the molar ratio of alkene to carboxylic acid produced in the oxidation reaction zone is in the range 0.8 : 1 to 1.4 : 1.

6. A process as claimed in claim 2 or claim 3 in which alkene and/or

carboxylic acid is separately recovered from the oxidation reaction product or separately added to the second reaction zone.

7. A process as claimed in claim 1, 2 or 3 in which the alkane is ethane, the corresponding alkene being ethylene and the corresponding carboxylic acid being acetic acid.

8. A process according to claim 2 wherein the alkyl carboxylate is ethyl acetate.

9. A process according to claim 3 wherein the alkenyl carboxylate is vinyl acetate.

10. A process according to claim 8 or claim 9 and wherein the molar ratio of alkene to carboxylic acid produced in the oxidation reaction zone is in the range 0.8: 1 to 1.4:1.

11. A process according to claim 1, 2 or 3 wherein the concentration of alkene fed to the oxidation reaction zone is from greater than 0 and up to and including 50 mol% of the total feed, including recycles.

12. A process according to claim 1, 2 or 3 wherein the concentration of water fed to the oxidation reaction zone is from 0 to 50 mol% inclusive of the total feed, including recycles.

13. A process according to claim 1, 2 or 3 wherein alkene and water are fed into the oxidation reaction zone.
14. A process according to claim 1, 2 or 3 wherein the alkene and water are fed into the oxidation reaction zone in an alkene : water ratio of 1 to 0.1-250 by weight.
15. A process according to claim 14 wherein the ratio of alkene : water is 1 to 0.1-10 by weight.
16. A process according to claim 15 wherein the molar ratio of alkene : carboxylic acid is in the range 0.8: 1 to 1.4 : 1.
17. A process according to claim 1, 2 or 3 in which the at least one catalyst in the oxidation reaction zone comprises molybdenum.
18. A process according to claim 3 in which the at least one catalyst in the second reaction zone comprises palladium.
19. A process according to claim 1, 2 or 3 in which the oxidation reaction is carried out at a temperature in the range 100 to 400 °C.
20. A process according to claim 1, 2 or 3 in which the oxidation reaction is carried out at atmospheric or superatmospheric pressure.

21. A process according to claim 1, 2 or 3 in which the oxidation reaction is carried out at a GHSV of 500-10,000 hr⁻¹.

22. A process according to claim 1, 2 or 3 wherein the product stream from the oxidation reaction zone also comprises carbon oxides in an amount of less than 15 mol%.

23. A process according to claim 1, 2 or 3 in which the alkane is ethane, the corresponding alkene is ethylene, the corresponding carboxylic acid is acetic acid and wherein ethylene and water are fed into the oxidation reaction zone in a ratio of 1 to 0.1-10 by weight, the molar ratio of ethylene to acetic acid produced is in the range 0.8: 1 to 1.4 : 1 and the product stream from the oxidation reaction zone also comprises carbon oxides in an amount of less than 15 mol%.

24. A process according to claim 1, 2 or 3, wherein water is introduced into the oxidation reaction zone.

25. A process according to claim 1, 2 or 3, wherein said adjusting or maintaining step is performed by controlling one or more of the pressure, temperature and residence time of the oxidation reaction zone.

26. A process according to claim 1, 2 or 3, wherein said contacting step is carried out in the presence of water and wherein said adjusting or maintaining step is

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performed by controlling one or more of the pressure, temperature and residence time of the oxidation reaction zone.

(IX) EVIDENCE APPENDIX

None.

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(X) RELATED PROCEEDINGS APPENDIX

None.